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EXPERIMENTAL UNRAVELING OF THE ELEMENTS AND ISOTOPES FORMED

IN URANIUM FISSION

From experiments by: Otto Hahn, Fritz Strassmann, and Hans Götte

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A series of elements of average atomic weight, usually in the form of several isotopes, originate in the fission of uranium; this was discovered by Hahn and Strassmann at the beginning of 1939. The number has grown to such an extent in the past three years that it is difficult for the layman to familiarize himself with the unraveling of the numerous substances. Substituents of 23 elements have been found to date. The number of analyzed isotopes is more than 80 as a result of their simultaneous appearance in various isotopes.

German   English

stab.   stable

kurz   [Note: For the figures   short

langleb.   1—19, see the appendix]   long-life

Illustration I.

Analyzed fission products of uranium fission

The problems which are to be solved here are more complicated than in the usual nuclear reactions. With the latter it is only necessary to analyze or separate isotopes of equal atomic number or their immediate neighbor elements. It is the chemist's task to find the fission products occurring, to arrange them properly with respect

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to each other, and to clarify the exact process of the decomposition series originating from the primary products to the stable isotope. We are far from having achieved a true arrangement of the primary isotopes into acceptable fission reactions. A number of short-life and long-life members of the individual series are also still missing. Illustration I shows results as they are known to us to date. (Several of the barium, lanthanum, strontium, and yttrium isotopes have not yet been described. They have been analyzed by Hahn and Strassmann.)

It should be noted that generally, in the case of synthetic isotopes, one always deals with the analysis of absolutely unweighable and invisible quantities of matter. These unweighable quantities of matter may be identified chemically just as exactly as the isotopes that are present in weighable quantities. During the discovery of uranium fission it was necessary to assume the presence of varium isotopes instead of the originally assumed radium isotopes. The assumptions generally valid at that time were thus nullified.

The usual chemical methods of analysis cannot be applied to the case of synthetically radioactive isotopes. The ionizing rays (β - or γ -rays) emitted by the radioactive isotopes are always used for analysis. In all cases one is limited to methods of counting the emitted particles, due to the weak available radiation sources in Germany. The simplest and most accurate method is the use of the Geiger-Muller counter, which has proved itself invaluable in the investigation of synthetic radioelements. The radiation particles emitted in a unit of time from the substance to be examined are

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registered in the Geiger-Muller counter. Conclusions are drawn
from the obtained activity curves concerning the simple or complex
nature of the measured substance, its half-life values, etc.

PRELIMINARY EXPERIMENTS GENERALLY TO BE UNDERTAKEN

Several examples will show the systematic clarification of
the processes.

Curve A of Illustration II shows the activity process of
a uranium preparation liberated from one of its natural transfor-
mation products, which was briefly exposed to a relatively weak
neutron source (Ra + Be). The straight rise is attributed to the
reformation of the β-radiating uranium X which was previously
separated from uranium. The initial decline shows the activity
process of the synthetic isotopes formed by the action of the neutrons
on the uranium. The chemical nature of these synthetic isotopes
is unknown. Curve B already indicates some progress. The separa-
tion of a group of synthetically active isotopes from uranium was
obtained by a hydrogen sulphide precipitation of a carrier element
capable of precipitation with hydrogen sulphide. The disturbing
reproduction of uranium X is eliminated.

Deflection
per
Minute

A - 10-minute exposure to radi-
ation, direct measurement

B - 30-minute exposure to radi-
ation, H_2S precipitation

Minutes

Illustration II

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Nothing of special significance may yet be recognized from Curve B. It must first be determined whether one is dealing with one homogeneous substance or a mixture of several. The fact is utilized that homogeneous radioactive isotopes decompose according to an exponential law, in such a fashion that in equal intervals of time the equal percentage of available quantities is transformed at any given time. If the measured activity is recorded logarithmically and the time arithmetically, a straight line is obtained for the decomposition of a homogeneous isotope. Curve A in Illustration III shows this for a hydrogen sulphide precipitate. A curved line is shown which gradually straightens. If the extrapolated values of the straight line are subtracted from the experimental values, a straight line is also obtained. Two different isotopes are apparently present. The longer one shows a half-life of about 60 minutes, and the shorter one of 16 minutes. (O. Hahn, L. Meitner, F. Strassman, Berichte d. Deutsch. Chem. Gesellsch. 69, 905, 1936.)

Actually there are additional isotopes present in the hydrogen sulphide precipitate. After very short exposure to radiation and immediate measurement a still more rapidly decreasing substance of about 2 minutes half-life appears in addition to the 16-minute substance. The long-life substance of about 60 minutes practically does not appear as yet. (O. Hahn, L. Meitner, F. Strassmann, Naturw. 26, 475, 1938.)

Intensity
on a

Log scale

Minutes

Illustration 3

Intensity

on a

Log scale

Minutes

Illustration 4

If the exposure to radiation lasts several hours or days a slower decrease of about 3 days^2 (Illustration 5) is observed in addition to the 60 minute half-life substances.

activity | UX (24 days)

3 day substance

days

Illustration 5

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After exposure to radiation for weeks or months a decrease of 60 days is observed (Illustration 6). (Hahn, Meitner, and Strassmann, Naturw. 26, 475, 1938.)

Counter deflections

per minute on a

log scale

$T_H = 60$ days

Time   Days

Illustration 6

The investigation of such long-life substances is relatively simple if these are present in sufficient intensity. They remain when the short-life substances have decomposed. This does not point to anything definite concerning the chemical nature of these substances, except that they belong to a group of elements capable of precipitation from an acid solution with H_2S, or that they originated from these elements after the precipitation. An exact chemical analysis is necessary in order to identify the synthetic isotopes.

CHEMICAL ANALYSIS OF AN APPARENT HOMOGENEOUS SUBSTANCE

One cannot assume from the approximate straight line decrease of a precipitation that a homogeneous isotope is present. Evidence

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obtained from the course of the exponential curves is not very
exact and isotopes of similar half-life values can easily simulate
a homogeneous decrease. A good example of this is given by close
chemical analysis of the aforementioned three-day substance obtained
from hydrogen sulphide precipitate. This so-called three-day sub-
stance proved to be still complex. It is a mixture of molybdenum
and tellurium. (Hahn and Strassmann, Naturw. 27, 451, 1939.) If
the solution of the three-day substance is mixed with small quantities
of molybdenum and tellurium and if these two elements are then sepa-
rated, the activity is distributed between both precipitates, and it
is readily seen that two different substances are involved. The
activity of the molybdenum decreases steadily within three days,
exactly 67 hours half-life, when the β-rays are measured in the usual
manner by means of 100 micron aluminum. The activity of the tellurium
initially increases for several hours and then decreases within three
days, (exactly 77 hours). (Illustration 7). In the later case a
transformation product is formed from the tellurium, which causes a
strong increase of intensity as a result of the penetrating β-rays.

Activity   the 2 three-day substances

on a
Log scale

Illustration 7

This daughter substance is an isotope of iodine, which decomposes with a half-life of 140 minutes. This iodine is separated easily from the tellurium and may be measured separately. (Illustration 8). The tellurium, on the other hand, increases correspondingly again. (Ph. Abelson, Phys. Rev. 55, 418, 1939; N. Feather and E. Bretscher, Nature 143, 516, 1939; Hahn and Strassmann, Naturw. 27, 451, 1939.) It is not possible to recognize the production of the iodine from the three-day tellurium during the investigation of the hydrogen sulphide precipitate, which contains the transformation products that decompose faster than three days (2-minute, 16-minute, 60-minute substances). The rapidly decreasing activities hide the increase of the iodine, so that only a separate investigation of the three-day substance resulted in clarification.

Activity

in

log scale

Iodine from three-day tellurium

Illustration 8

The close investigation of the 60-day substance, which is contained in the H_2S precipitate after long exposure to radiation, is analogous. It is necessary to await the decomposition of all shorter life substances of the H_2S group. Here, too, a chemical analysis shows that the 60-day substance is also not a homogeneous

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isotope, but a mixture of isotopes of various chemical elements.

The analysis of the short-life substances, for example the mentioned two-minute substance, may be much more difficult. The satisfactory precipitation and purification of a hydrogen sulphide precipitate takes at least several minutes. After this the various chemical separations begin. In the case of the hydrogen sulphide group, these require a somewhat ample length of time. A number of elements can be excluded, therefore, in the case of the two-minute substances. However, a true allocation has not been achieved to date.

A MORE EXACT ANALYSIS OF THE STRONTIUM ISOTOPES AS AN EXAMPLE OF SYSTEMATIC PROCEDURE

A single group of elements is selected for a more exact analysis.

The alkali earth isotopes, barium and strontium, are an important and isotope-rich group of isotopes necessary for the fission of uranium. Representatives of the alkaline-earth metal, calcium, do not seem to appear. Unraveling the individual half-life values from the decay curves of the active barium and strontium isotopes, which were precipitated together, would yield poor results. The task is simplified by the easy and rapid separation of both barium and strontium from uranium, uranium X, and all other fission products.

Activity

Strontium I = 7 minutes

Illustration 9

The strontium group is selected for our considerations. In the case of barium the conditions are similar, although more difficult for the non-chemist.

The method of investigation is again very similar to that of the hydrogen sulphide group. With strontium, however, we are dealing with only a single element. An approximate estimate of the expected results is obtained by exposing uranium to radiation for various lengths of time. This is followed by rapid rectification of the synthetic strontium isotopes, by separating and reprcipitating them, using strontium salt as the activity carrier. Three strontium isotopes of varying half-life values were easily determined. (Cl. Lieber, Naturw. 27, 421, 1939). Illustration 9 shows the activity process of a strontium preparation which was separated after exposing uranium for 16 minutes to radiation. A very rapid initial decrease changes to a weakly marked, very gradual rise. A flat maximum is reached after $3\frac{1}{2}$ to 4 hours, after which a gradual decrease begins. Curve c

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is obtained by extrapolating the weakly rising curve b to $T = 0$,
and subtracting the obtained values from the experimental curve.
Curve c shows an exponential decrease of 7 minutes half-life, which
doubtlessly belongs to a strontium isotope. Curve b may stem from
a second strontium isotope, which forms an active metabolon (yttrium)
during its decomposition. The curve could also be attributed to yt-
trium and a zirconium formed from this yttrium.

Activity

Illustration 10

Illustration 10 shows that the first assumption is correct.
The uranium was exposed to radiation for several hours. Curve (a)
shows the decay after immediate separation of the strontium (much
smaller scale than Illustration 9). The strontium precipitation in
curve 9 was undertaken about one hour after the interruption of
radiation. The 7-minute strontium isotope disappeared. But considera-
ble increasing activity, weak during the first few hours, is still
present in the strontium precipitate. A second strontium isotope
is present, which reproduces an yttrium of shorter half-life, and
then decomposes with its half-life in equilibrium with the yttrium.
Illustration 11 shows the decay of such an yttrium as proof for the

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validity of our conclusion. The half-life of the yttrium is 3.5 hours. The half-life of its mother substance shows a straight line decrease according to Illustration 10. This decrease apparently belongs to a strontium with a 6-hour half-life (Cl. Lieber, Natur-wissensch. 1. c.)

Activity

Minutes

Illustration 11

The 55-day strontium

Activity

54 days

54 days

57 days

Illustration 12

It was now necessary to show whether another strontium iso-tope appears after longer exposure to radiation, which may have es-caped detection during shorter exposure to radiation. This is indeed

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the case. Curves (a) and (b) of Illustration 12 show several decay curves which were obtained therefrom. The rapid decay of the 6-hour strontium is followed by a slower decay of approximately 54 to 55 days half-life. The transition of the initially rapid decay into the straight line decay with 55-day half-life is very noticeable, and will be reverted to later.

Curve (c) and (d) of Illustration 12 show that the 55-day substance is strontium. The final strontium precipitations were undertaken after the shorter life isotopes decomposed quantitatively. The decay corresponds well with a decomposition period of 55 ± 3 days with some initial irregularities. (see footnote 1, page 10).

The conditions thus far are clear. Three isotopes, 7 minutes, 6 hours, and approximately 55 days, were determined. The 6-hour substance reproduces yttrium of 3.5 hours. Actually the situation is much more complicated.
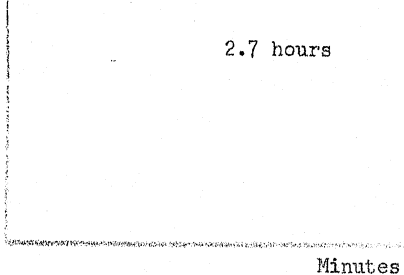
Further clarification was achieved by means of stronger radiation sources. Using weaker sources, much longer and continued exposure to radiation of uranium over periods of months was also undertaken. The 6-hour isotope as well as the long-life isotope of strontium are still complex.

We now turn to the 6-hour strontium. It was earlier noted that the increase was small only as a result of the reproduction of the 3.5-hour yttrium (compare illustrations 9 and 10). This is explained as a result of a relatively strong absorbing β-radiation

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of the yttrium, which participates only weakly in the 100-micron counter. An absorption curve of these β-rays shows that highly penetrating rays are present. The weak increase of the 6-hour strontium is explained by assuming that the 6-hour substance consists of more than one isotope, from which only the one 3.5-hour yttrium is reproduced. (H. Götte, Naturw. 29, 496, 1941.) To prove this assumption a uranium preparation was exposed to radiation by means of neutrons from the high tension apparatus of the Royal Scientific Institute for Physics, instead of using Ra-Be tubes. Then the strontium was separated and purified, and the reproduced yttrium quantitatively separated and measured every two hours. A set of curves was obtained whose initial activity decreased in a manner similar to the mother substance of the separated yttrium. This decrease took place with a half-life of 2.7 hours.

Illustration 13, curves A (1-9) and curve B show the results. The 3.5-hour yttrium has as mother substance a strontium isotope with a 2.7 hour half-life. The earlier found half-life of 6 hours must stem from at least two isotopes. One of these decreases with a 2.7-hour half-life and the other with one of more than 6 hours. The combined action of these two isotopes simulates the 6-hour decrease. This longer isotope was also found. It has a half-life of 8.5 hours. It is easily detected from strong preparations by completely decaying the 2.7-hour strontium and then separating and measuring the remaining long-life strontium from the yttrium which formed in the preparation.

2.7 hours

Minutes

Illustration 13

Curves are obtained which stem from the 8.5-hour and the
above-mentioned 55-day strontium. At the same time it was deter-
mined that the 8.5-hour substances is not only the mother substance
of an earlier found 57-day yttrium (O. Hahn, and F. Strassmann,
Naturw. 28, 543, 1940.), but that at the same time another yttrium
isotope with a half-life of 50 minutes is produced from it.
(see Illustrations 14 and 14 a). It could not yet be decided whether
two isomeric yttrium isotopes were produced from the one strontium iso-
tope with a half-life of 8.5 hours, or whether the 8.5-hour sub-
stance itself consists of two isotopes with very similar half-life
periods from which the one forms the 57-day substance and the other
the 50-minute substance. (For further details concerning these
experiments see H. Götte; see above).

A number of decay curves for active strontium were indicated
in Illustration 12. This strontium was separated from uranium which
was exposed to radiation for several days. These curves showed certain
small anomalies in that a very minute increase of activity was always

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observed during the transition to the 55-day decay. This increase
was outside the margin of experimental error. The yttrium was again
separated from the decreasing strontium with a 55-day half-life.
This yttrium always showed a very minute but definitely detectable
activity. This weak, separable activity was not similar to the
55-day decrease of its assumed mother substance.

hours

8.5 hours

minutes

Illustration 14

hours

8.6 hours

50 minutes

minutes

Illustration 14a

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The possibility had to be considered that the 55-day strontium was still complex and that a longer life strontium was present in smaller quantity, which was the mother substance of the very weak, active yttrium.

The unknown, longer life strontium should be obtained in somewhat larger yields when uranium is exposed to radiation over a period of many months. This assumption was verified. The half-life of a strontium separated from a uranium that was exposed to radiation for 11 months, gradually lengthened to more than 55 days. From such a strontium an yttrium isotope may be separated from time to time, the initial intensity of which decreases very slowly. Illustration 15 shows two experiments performed according to the indicated method. Curve (a) shows the yttrium separated from the strontium, after the decrease of the so-called 55-day strontium was observed for one month. After the separation of the yttrium, the strontium was measured in the same counter and the same arrangement, as before. The strongly decreasing activity increases again after several days. The decrease continues, but the decay becomes gradually slower, according to the half-life of 55 days (dotted line).

Analysis of the complex nature of the 55-day strontium

1. y-separation

Activity

y-
separation

2. y-separation

Days

Illustration 15

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Curve (b) readily indicates this. The yttrium was separated for the first time after 80 days (Point 1), and measurement of the strontium was continued. The decay becomes constantly slower. 570 days after the start of the measurement the yttrium was separated again with the relatively weak preparation. (Point 2 of curve (b)). The obtained strontium now shows an activity increase much clearer than the previous increase. After this time the 55-day strontium decomposes to less than one thousandth of its initial activity. Therefore, the strontium isotope must be a new one of very long life which reproduces an active yttrium. We judge the half-life of this yttrium isotope to be about 60 hours. These experiments thus prove that the 55-day strontium is complex and that it contains a smaller or larger percentage of a long-life isotope depending upon the length of time the uranium is exposed to radiation. Its half-life could not be exactly determined. It probably is more than two years, and that of its transformation product about 60 hours.

The pure 55-day strontium is obviously identical to an active isotope of atomic weight 89, which was earlier obtained from the strontium itself. This isotope changes into the stable yttrium of the same atomic weight. The remaining strontium isotopes form active yttrium isotopes, which in turn transform into zirconium.

At first it was not possible to determine an active yttrium for the 7-minute strontium. Recently this determination was made possible by the much stronger neutron source of the Max-Planck-Institute for Physics.

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With preparations available until recently, the search was almost hopeless for a presumed, less transformation product of the 7-minute isotope. Too short an exposure to radiation was not feasible, since the intensity of the strontium would not have been sufficiently great. For the same reason considerable quantities of uranium were used for radiation. However, the separation and purification of the strontium took so long, that the 7-minute substance largely decomposed and the isotopes with a longer life interfered.

With the stronger neutron source the duration of radiation was reduced to a few minutes. By means of a simple procedure large quantities of uranium could be used rapidly and almost at will to treat the sought isotope. For this purpose, for example, 50 grams of uranium nitrate were separated from uranium X before exposure to radiation by repeated agitation of its ether solution with small quantities of water. The ether-uranium solution containing a few drops of water was now exposed to radiation. The agitation took place en route from the radiation to the processing location. The small quantity of water contains in addition to a small amount of uranium the larger part of all the fission products. Among these is strontium, which may now be easily separated from the remaining fission products and also from the different barium isotopes, which are chemically close to strontium.

The decay of this strontium after exposure to radiation of four minutes takes place similarly to the strontium curve of Illustration 9. The short, strong exposure causes the activity of the 7-minute strontium to be delayed in relation to its longer life

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isotope. It could be hoped that it was possible to determine an
active yttrium, originating from the strontium, in addition to the
3.5-hour yttrium. Therefore, the yttrium was separated from the
strontium 20 minutes after the separation of the strontium, and its
activity progress was measured. After an insignificant, initially
rapid decrease a straight line resulted, which corresponded to a
half-life of approximately 9 hours.

If the strontium was permitted to stand for 1 hour and was
then separated from the yttrium, and if this was succeeded by another
yttrium precipitation from the strontium after two hours, the stron-
tium decreased considerably faster. The half-life was about 3.5
hours, so far as could be determined with the small activity. This
was verified by renewed strong exposure of uranium to radiation for
16 minutes. During this time a larger percentage of the 2.7-hour
strontium is formed, and from this a correspondingly greater amount
of yttrium of 3.5-hour half-life. After allowing the separated stron-
tium to stand for 20 minutes, the existence of the new 9-hour
yttrium was definitely recognized. Yttrium precipitated from the
aged strontium, after an intermediate precipitation, contained
only the 3.5-hour yttrium.

Origin of an 8-hour yttrium from a 7-minute strontium

Activity | 3.5 hours
            |
            | 8 hours
            | 3.5 hours
            | 8 hours
            |_____
            Hours

Illustration 16

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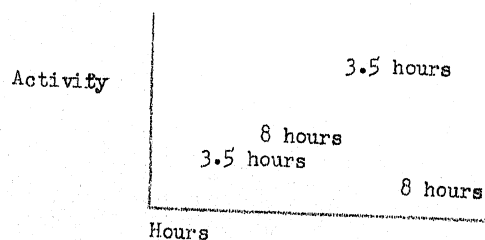
Illustration 16 shows these results. Curve (a) indicates the decay of the yttrium separated from the strontium after 20 minutes in the case of the four-minute experiment. Curve (b) shows the later yttrium precipitation. Curves (c) and (d) show the results of the strontium obtained during the longer exposure to radiation. The initially more rapid decrease of the first yttrium curve (c) is more marked than in the case of (a), because of the presence of a large percentage of the long-life strontium isotope.

These experiments force us to conclude that the 9-hour yttrium originates from the short-life strontium isotope. If the latter is decomposed, which occurs after one hour, and if the yttrium originating therefrom is removed, then only the 3.5-hour yttrium and traces of the 57-day and 50-minute yttrium isomers are formed from the remaining, more stable strontium isotopes.

When the 9-hour activity of curve (c) is extrapolated to $T = 0$ and the resulting values are subtracted from the measured ones, one does not obtain a true 3.5-hour curve, but an initially faster decay. This perhaps indicates the existence of another still unknown yttrium isotope, which may also indicate the complex nature of the 7-minute strontium.

The 9-hour yttrium isotope is missing in Illustration 13. This is caused by the isolation of the strontium, from which the yttrium fractions were precipitated, after the complete decomposition of the short-life strontium isotope.

The strontium isotopes were used as an example to describe

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the various methods for analyzing and characterizing the five dif-
ferent active isotopes and their first transformation products, the
yttrium isotopes.

These experiments do not yet indicate whether these strontium
isotopes are primary fission products or transformation products
of elements of lower atomic number. Since an entire series of barium
isotopes was found during the fission of uranium, it was assumed
that the appearance of this group indicated a simultaneous origination
of strontium and xenon on the one hand, and barium and krypton on
the other. Both element pairs with the atomic numbers 38 + 54 and
56 + 36 add up to 92, which is the nuclear charge of the splitting
uranium. In one of the earliest reports on nuclear fission the forma-
tion of xenon was indicated (O. Hahn and F. Strassmann, Naturw.
27, 163, 1939). Shortly thereafter Dutch (F. A. Heyn, A. H. W.
Aten, C. J. Bakker, Nature 143, 516, 679, 1939) and American
(A. Langsdorf, jr., Phys. Rev. 56, 205, 1939) scientists, and we our-
selves demonstrated the formation of krypton. (O. Hahn and F. Strass-
mann, Naturwissenschaften 27, 529, 1939.) It was necessary to
determine which of the strontium isotopes described above are
possible transformation products of krypton, and which do not stem
from a gas.

The method for showing the existence of rare gasses original-
ly used by us was as follows: A stream of air was led through the
uranium solution during exposure to radiation, causing the sought
active rare gas to be led over cooled activated charcoal. The char-
coal was then converted into alkali or alkaline earth metals. If

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such were found, they could only have originated from an active rare
gas in the charcoal, since the metals or their salts would not pass
over with the air stream. In this manner some cesium, barium, and
rubidium isotopes were detected.

With weak radiation sources active strontium isotopes were
found by means of a new method. Use was made of the "emanating ability"
of surface rich uranium compounds for rapid detection of active rare
gases. (O. Hahn and F. Strassmann, Naturw. 28, 54, 1940.) The
rare gases originating during radiation diffuse outwardly from the
"Highly emanating" uranium preparation. Transformation products
originating from the active gases may be collected on a negatively
charged metal plate. The "active precipitates" may be removed from
the plate in fractions of a minute, and may then be chemically sepa-
rated from one another. In this manner, in cesium and rubidium isotopes
and several barium isotopes, as well as the 2.7-hour, the 8.5-hour and
the 55-day strontium were determined as derivatives of krypton isot-
opes. (O. Hahn and F. Strassmann, Naturw. 28, 54, 1940), (H. Gütte,
see above). It may be added that in later experiments a direct
separation of the xenon and krypton isotopes and their decomposition
products was achieved by directing the air stream through the
uranium solution. Charcoal adsorption containers, placed behind
one another, were used. One was cooled with dry ice + alcohol, and
the other with liquid air. The xenon was readily adsorbed by means
of the dry ice and the krypton by means of liquid air. (O. Hahn and
F. Strassmann, Naturw. 28, 455, 1940.)

It was also possible to determine the 7-minute strontium as

a krypton derivative by means of the strong neutron source of the Max-Planck-Institute. The detection from the "highly emanating" uranium was not possible earlier. Apparently the 7-minute strontium yields were too small in comparison to the longer life isotopes, even with the shortest permissible radiation time for a co rect measurement. From this it is concluded that the krypton isotope forming the 7-minute strontium possesses an especially short half-life. In agreement with the above-described analysis of the origin of an yttrium isotope of 9-hour half-life from the 7-minute strontium, this new yttrium isotope was found in the "active precipitate" from "highly emanating" uranium.

It has not yet been determined whether the long life strontium of more than two years also originates from krypton.

The result of the described analysis of the strontium isotopes found during the fission or uranium, their mother substances, and metabolons is represented in the following diagram:

$$36^{Kr} \qquad 37^{Rb} \qquad 38^{Sr} \qquad 39^{Y} \qquad 40^{Zr}$$

? ⟶ ? ⟶ 7 minutes ⟶9 hours ⟶?

very short → 80 seconds ⟶ 2.7 hours ⟶3.5 hours ⟶ stable

? ⟶ ? ⟶ 8.5 hours ⟨ 57 days / 50 minutes ⟶ ?

2.5 minutes →15.4 minutes ⟶55 days ⟶ stable
⟶ 2 years ⟶ 60 hours ⟶ stable
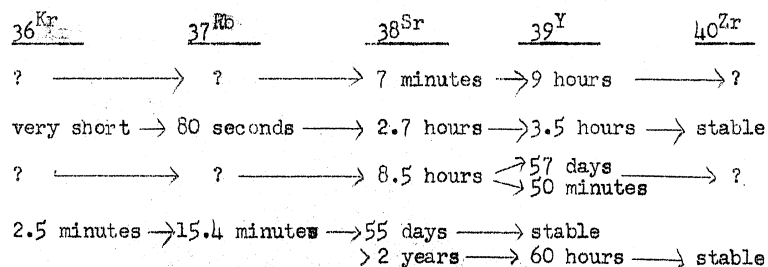
Only the chemical unraveling of the strontium isotopes, their origin and transformation were dealt with. Therefore, the determination of the half-life of the krypton and rubidium isotopes, their

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probable atomic weights, etc., was not discussed. (Further details
concerning the krypton-strontium group see: W. Seelmann-Eggebert,
Naturw. 28, 451, 1940; H. Götte, Naturw. 28, 496, 1940; O. Hahn and
F Strassman, Naturw. 28, 455, 1940.) A discussion concerning the
atomic weightsof all the fission products, so far as they are
determinable, will be undertaken in another report written in col-
laboration with Professor Mattauch.

PHYSICAL CHANGES OF THE WORKING REQUIREMENTS AS AN AID
TO THE EXPERIMENT

It is not the intention of this report to discuss the other
elements and element groups appearing during nuclear fission in
a manner similar to that in which strontium was discussed. This
would be a repetition of all chemical processes applied in this field.
However, it may be of interest to present some examples which show
the significance of systematic changes in the method of exposing
uranium to radiation and the measuring of the fission products.

1. Duration of Exposure

An important point is the duration of exposure. The almost
hopelessly entangled mixture of different isotopes and their trans-
formation products is divided into distinct groups by exact balancing
of the length of exposure and by using the individual separations
at different times after the exposure. This processing method has
already been used in the preliminary tests as well as in the case
of the strontium, as previously discussed.

2. Energy of the Reacting Neutrons

The previously exemplified fission processes, as well as all
others investigated at the Kaiser-Wilhelm Institute, are best re-
leased by means of slowed neutrons. The radium-beryllium tubes,

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which were used exclusively until recently as a neutron source were surrounded by paraffin. The preparations which were to be exposed were located several centimeters away from the tubes. Other thick paraffin layers were placed behind the preparations. If the paraffin is omitted from the same geometrical arrangement, the fission product yield will be much smaller. Thermal neutrons are the most effective. However, the chemical nature of the fission products was always the same even when the most energy-rich neutrons were used from the radium beryllium preparations.

This is changed when the energy-rich neutrons of the Li-D-reactions, (action of deuterons upon Li in high tension apparatus) which amount to 17 million electron-volts are used for exposure. It was first determined by Japanese scientists (T. Yasaki, Sc. Pap. Inst. Physic. chem. Res. Japan 37, 457, 1940. Y. Nishina, T. Yasaki, K. Kumura and M. Ikawa, Phys. Rev. 58, 660, 1940, 59, 323, 677, 1941. Nature 146, 24, 1940) and verified by Americans (E. Segre and G. T. Seaborg, Phys. Rev. 59, 212, 1941) that completely new fission processes result from these energy-rich neutrons. Isotopes of the elements ruthenium (44), rhodium (45), palladium (46), silver (47), cadmium (48), and indium (49) originate. The fission processes proceed in this case more symmetrically than with slow neutrons. In addition to these symmetrical fissions, the remaining known fission processes are undoubtedly released, since slower neutrons occur in considerable quantities in the spectrum of the Li-D neutrons. We found the yield of symmetric to known fissions to be

in a ratio of 1:1, during investigations concerning the appearance
of the symmetrical fissions during Li-D-exposures and the covering
of the neutrons with cadmium and borium. If exposures are undertaken
with unslowed Li-D-neutrons, the appearance of all the fission ele-
ments during the chemical analysis must be considered.

During the exposure of uranium, so-called resonance processes
occur. Neutrons possessing a certain energy are added to the uranium
(n, γ-process) (L. Meitner, O. Hahn and F. Strassmann, ZS. f. Phys.
106, 249, 1937.) or to processes during which a second neutron
leaves the uranium nucleus (n, 2n-process) (Y. Nishina, T. Yasaki,
K. Kumura and M. Ikawa, Phys. Rev. 57, 1182, 1940; T. Yasaki, Sci.
Pap. (see above); E. McMillan, Phys. Rev. 58, 178, 1940) together
with the originally radiated neutrons. In the first case the uranium
isotope of 23-minute half-life is formed already before detection
of the uranium fission (E. McMillan, Phys. Rev. 58, 178, 1940). In
the latter case a 7-day isotope is formed (O. Hahn, L. Meitner, F.
Strassmann, Ber. d. Dtsch. Chem. Ges. 69, 912, 1936.). An isotope
of element 93 with a half-life of 2.3 days is formed from the 23-
minute substance (E. McMillan, P. H. Abelson, Phys. Rev. 57, 1185,
1940). The possible presence of these substances must also be
considered, if the individual fission products are to be obtained in
a pure form.

3. Strength of exposure

It is almost too trivial to mention that a stronger exposure of
the uranium and the resulting greater yield of fission products
has advantages as opposed to weaker preparations. It is, however,
instructive to show by means of an example, how the investigation
may be made which could not be made with weak preparations. Barium

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is selected as an example. Three isotopes had previously been de-
termined, one of 14 minutes, one of 86 minutes, and one of about 300
hours half-life. The 14-minute and the 300-hour isotopes still give
active lanthanum isotopes. The half-life for the first lanthanum
was judged to be about $2\frac{1}{2}$ hours, while that for the latter was de-
termined at 44 hours (O.Hahn and F. Strassmann, Naturw. 28, 543,
1940.) It was suspected that the 14-minute barium might be a
primary fission product. The origin of the other two was definitely
traced to xenon. (O. Hahn and F. Strassmann, Naturw. 27, 529, 1939.)

Results concerning the 14-minute barium have essentially
changed due to the stronger preparations resulting from use of the
high tension apparatus of the Max-Planck Institute. The 14-minute
barium is still complex and consists of two isotopes which them
selves reproduce two active lanthanum isotopes. This cannot be
detected with the usual decay measurement. The 86-minute isotope
which is simultaneously present together with the 14-minute barium,
and the lanthanum which is repreduced from the barium make the decom-
position curve too complicated to permit a true evaluation. The
following procedure was followed when the strong neutron source of
the high tension apparatus was used. The exposure lasted only a
few minutes. This causes the production of only small quantities of
86-minute barium, since it first forms from a 7-minute cesium iso-
tope and a 45-second xenon (see table in Illustration 1).

The uranium was processed into barium as quickly as possible
immediately after its exposure. The barium was dissolved and the
solution increased to a larger volume. A certain, gradually increased
number of cubic centimeters of this stock solution were mixed with

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several milligrams of iron during intervals of six minutes each.
The lanthanum, which reformed in the meantime, was separated from
barium by means of this iron. The barium was quantitatively pre-
cipitated and measured twice for two minutes at a time. Each time
the activity of the barium isotopes still present were obtained
free from the disturbing lanthanum. By systematically increasing the
precipitated quantity of active barium from the stock solution and
adding each time the same quantity of inactive barium as a carrier,
the activity measured beyond four minutes may always be kept app-
roximately equal. Thus the accuracy is not affected by the constantly
weakening active barium. The direct decay of the pure barium iso
topes, free from decomposition products, is obtained by always re-
lating the measured activities to the same volumes of stock solu-
tion. It was shown that the decay of the so-called 14-minute barium
did not correspond to a homogeneous substance (the decay is corrected
to the small quantity of 86-minute barium). A curved line was obtained
whose evaluation points to the presence of a 6-minute and an 18-min-
ute decomposing barium. (Details concerning this follow in a re-
port soon to appear by Hahn and Strassmann).

The lanthanum originating from the barium isotopes was care-
fully investigated in another series of experiments. Desired
results are obtained when the lanthanum separations are undertaken
while the transformation products of both barium isotopes are still
present. In another method the lanthanum isotopes are separated
after the decomposition of the 6-minute barium; lanthanum is produced
from the remaining 18-minute barium after a certain waiting period,

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and then measured. The experiments are still not completed. Each
of the two short-life barium isotopes reproduces an active lanthanum
isotope. The half-life values are approximately 70 minutes and 3.5
hours. (Details concerning this follow in a report soon to appear
by Hahn and Strassman).

Another result stemming from the use of stronger prepara-
tions is the evidence that the two short-life barium isotopes must
also originate from xenon. Both were found on the negatively charged
plate with the help of the "highly emanating" uranium. The fact
that the 14-minute barium, now recognized as complex, could formerly
not be shown as the derivative of xenon, again points to short-life
xenon isotopes, as in the case of the 7-minute strontium described
above. Only a small part of the short-life xenon isotopes diffuses
out of the uranium before it decomposes.

The example used to point out the advantages of stronger
radiation sources of the now recognized complex 14-minute barium,
may also be applied to isotopes previously considered homogeneous.
Doubtlessly, other fission products will be found with sufficient
radiation intensity. Some of them originate partly as so-called
secondary reactions, as do two of the bromium isotopes found by us.
(O. Hahn and F. Strassmann, Naturw. 27, 529, 1939; Naturw. 28, 817,
1940.) Also other reactions enter, whose intensity is too small to
make their detection possible. Such secondary reactions are defi-
nitely important for the complete solution of the fission processes.

4. Hardness of Radiation Used for Investigation

Of great importance during the investigation of individual fission products is the testing of the substances with multiple layers of absorbing metal foils, and their comparison in counters of normal and very small thicknesses. By interposing absorbing foils, those isotopes obtained from mixtures of various isotopes, and which emit the most penetrating β-rays, are first measured. Isotopes heretofore not detected, were obtained by this method. Measurement of the absorbable β-rays is made more feasible by the use of counters with especially small thicknesses, for example 5-micron instead of the usual 100-micron aluminum. It is possible, with the smaller thicknesses to detect isotopes which appear only weakly or not at all with use of the 100-micron counter.

A few examples will be given for both methods of investigation. An 18-minute molybdenum isotope, detected recently during the fission of uranium, made possible the determination of a 14-minute isotope, as a transformation product, from element 43. (O. Hahn and F. Strassmann, Naturw. 29, 369, 1941; ZS. f. Physik 117, 789, 1941.) The decay of the molybdenum occurred in form of a straight line, and was not dependent on whether element 43 was present or had been separated shortly before. This is explained by the fact that the mentioned molybdenum still consists of two isotopes. One of these is the mother substance of the 14-minute isotope, the reproduction of which results in an initial increase. The other one decreases initially, thus compensating for the increase of the first one. After a number of futile attempts, molybdenum isotopes

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freshly obtained from uranium fission were measured through aluminum
foils of various thicknesses. It was shown that the measured half-
life values decreased gradually from 18 minutes to 12 minutes
with increasing thickness of the absorbing layers. Use of additional
foils produced no further change in this value (Illustration 17).
This proved the existence of a new 12-minute molybdenum isotope.
The 12-minute molybdenum isotope cannot be confused with the 14-
minute isotope of element 43, since the reproduction of this meta-
bolon would have evidenced a delay -- but no reduction -- of the
18-minute decrease. (For details concerning these very complicated
procedures with molybdenum see W. Maurer and W. Ramm, Naturw. 29,
368, 1941; and Hahn and Strassmann, l. c. Naturw. and ZS. F. Phys.,
l c.) In addition the β-rays of element 43 with 14-minute half-
life are completely absorbed by two to three millimeters of alumi-
num.

Analysis of the complex nature of active molybdenum

Activity

minutes ⟶

Illustration 17

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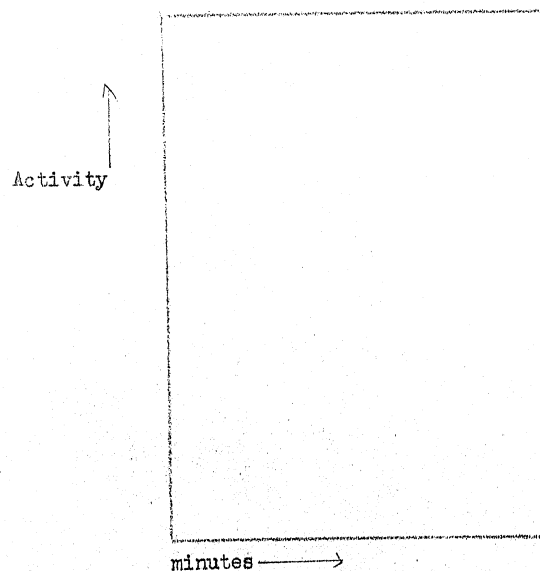
Some additional examples will be given to show the advantages
of a very thin walled counter for the measurement of highly absorb-
able rays. It has already been pointed out that during the exposure
or uranium to neutrons a resonance process and a (n-2n) process
occur in addition to the actual nuclear fission. These lead to
synthetically active isotopes of the uranium itself. One of these
isotopes is the well-known decomposing 23-minute substance, originating
from an n, γ-process. (O. Hahn, L. Meitner and F. Strassmann, Ber.
d. Dtsch. Chem. Ges., l. c.). As American scientists have demonstrated,
an active isotope of element 93 originates during the transformation
of this substance, and decomposes with a half-life of 2.3 days.
(McMillan and Abelson, Phys. Rev., l. c.).

The other uranium isotope originates with very fast neutrons
according to an n, 2n-process, as pointed out earlier. It was first
discovered by Japanese scientists and verified by American scien-
tists. [(Y. Nishina, T. Yasaki, K. Kimura and M. Ikawa, Phys. Rev.
57, 1182, 1940; t. Yasaki, Sci. Pap., l. c.) (E. McMillan, Phys. Rev.
58, 178, 1940.)]. It has a half-life of 7.0 days. According to the
experiments of the above scientists, the 2.3-day substance and the
7-day uranium isotope do not emit very penetrating β-rays. Accord-
ing to Starke this is also true of the 2.3-day substance. (K. Starke,
Naturwissensch. 20, 107, 1942.) Its detection is therefore not very
easy in the normal 100-micron counter, and requires at least considera-
ble activities. Here the advantages of the thin-walled counter as
compared to the 100-micron counter, and even more so in comparison
to aluminum added in series, are apparent. The illustration shows

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the decay of a mixture of the 23-minute uranium with the 2.3-day substance with equal radiation intensity, when measured in a 5-micron, a 100-micron, and finally with an added 500-micron aluminum counter. The preparations used for the three measurements were taken from the same stock solution, which naturally contained some ordinary uranium. About twice as much was used for testing through the 100-micron counter than was used in the 5-micron counter. For the added 500-micron counter considerably more was used. The weak activity of the uranium, or the reproduced uranium X, was obtained for each test and was sub-tracted from the the experimental values. For the comparison of the radiation absorbtion, the measurement results were related to equal quantities of initial substance (Illustration 18). Curve A. of the illustration (measurement by 5-micron aluminum) slopes immediately after the beginning of the measurement. It s clearly seen that a mixture of equal parts of the two substances is present. In curve B, the 23-minute substance predominates. The 23-minute substance is decomposed after approximately 200 minutes. The ratio of the activities of curves A and B after this time is 6:1. This is proof that the rays of the 2.8-day substance are much more absorbable than those of the 23-minute uranium isotope. The weak β-rays of the 2.3-day substance are barely demonstrated by curve (c). The true 23-minute decrease of the uranium isotope is obtained.

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Suppression of rays of the 2.3-day
substance in a mixture with the 23-
minute uranium by addition of alu-
minum foils

Absorption measurements
of element 93

Activity

Minutes

Illustration 18

The rays of the 7-day uranium isotope are still more ab-
sorbable than those of the 2.3-day substance. The β-rays of
the 7-day uranium isotope are hardly detectable in the normal
100-micron counter if very high intensities are not available.

The 5-micron counter easily permits recognition of this
substance, and its decay may be determined. The 7-day uranium is
precipitated simultaneously with the ordinary uranium. Therefore,
a decay curve plotted over a period of weeks using the same precipi-
tate would indicate very uncertain values because of the unavaidable
reproduction of uranium X. As in other cases, a certain amount

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was precipitated at regular intervals as uranium sodium acetate
from a stock solution of the 7-day substance plus the uranium.
This removes uranium X. Exactly-measured quantities of the precipi-
tate (four samples each time) were taken successively in a 5-micron
counter. The α-rays of the uranium were made harmless by a sufficiently
thick layer of cellophane. The scattered values resulting from the
somewhat changing layers, are balanced by the four measuring
points, at least in the beginning of the curve, where the intensity
of the 7-day substance is still rather strong. The illustration
shows the result of such a series of measurements (Illustra on 19).
The straight line is drawn through the points with a half-life of
7 days. It is in good agreement with the obtained activities. The
results of the Japanese and American scientists are herewith well
verified.

Although the briefly-discussed uranium isotope of 7-day
and element 93 of 2.3-day half life are not uranium fission
products, they are mentioned in our considerations concerning the
unraveling of the fission products. These isotopes which originate
during exposure must be considered during the individual separations.
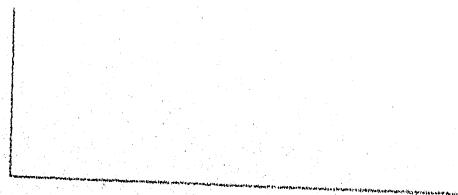
Illustration 19

The 7-day uranium isotope

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CONCLUSION

The physical changes discussed under 1 to 4 that were involved
in the practical investigation of fission reactions, must not be con-
strued as complete. They only indicate that which proved to be ad-
vantageous during the chemical analysis with our limited equipment.
Varied methods for more exact insights into the processes are avail-
able to the physicist. Investigation of the rays according to the
Wilson cloud-chamber method, where direct evidence concerning the
energy of the investigated β-rays and their electric charge is
given, may be mentioned. The nature of the charge, whether positive
or negative, is also obtained in counters with a series-connected
magnetic field. The control of the charge indication has proved to
be advantageous for the determination of whether or not a fission
product is identical to a synthetic isotope already described and
obtained in the usual manner. (W. Bothe and A. Flammersfeld, Naturw.
29, 194, 1941.) The β-ray spectrometer may be used for a more exact
investigation of individual radiation groups. The energy distribu-
tion of the β-rays emitted from the individual isotopes is determined
by a method other than the direct absorption measurement. This leads
to a method for determining whether the obtained electrons are nuc-
lear β-rays, thus indicating an element of higher atomic number,
or whether an activated atom transforms into the ground state of the
same atom, thus emitting electrons of discrete energies, i.e., an
electron-line spectrum, as a result of so-called internal transfor-
conversion
mation. If the characteristic X-rays of the atom, released during

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nuclear processes, are investigated, the atomic number of the metabolon may be determined, and, for example, the presence of a K-electron capture may also be determined.

The possibilities of physical investigation just mentioned, which proved very successful, especially in America with its strong radiation sources, were not discussed in detail, because only a survey concerning chemical procedures was intended. For complete knowledge of all processes the cooperation of physics and chemistry is a necessity. It is to be hoped that also in Germany the possibilities for varied investigations will be utilized after the construction of more and stronger radiation sources.

E N D

Appendix

$$^{235}_{92}U + ^{1}_{0}n \qquad ^{238}_{92}U + ^{1}_{0}n \qquad ^{235}_{92}U + ^{1}_{0}n$$

Figure 1. Analyzed fission-products of uranium fission.

A

deflection per minute

A: 10-min irradiation, direct measurement
B: 30-min irradiation, H_2S precipitation

Figure 2.

intensity (on logarithmic scale)

Figure 3.

B

intensity (on log scale)

A
N
A-N
B
$T_H = 16$ min
$T_H = 3.2$ min
$T_H = 2.1$ min

400
200
100
40
20
10

5   15
17   27   41   57   minutes

Figure 4

activity

100
50
20
10
5

UX (24 days)
3-day substance

0   2   4   6   8   10   12   days

Figure 5

counter
deflections/minute
(on log scale)

A
$T_H = 60$ days
B
$T_H = 60$ days

Figure 6

0   100   200   time (days)

C

activity (on log scale)

the 3-day substances

Figure 7

iodine
from
3-day
tellurium
129'
139'   136'

Figure 8

120   240   360   480   600   720 minutes

activity

Figure 9

strontium
I = 7 min
(c)

(a)

(b)

40   80   120   160   200   240   280   0 20

D

Figure 10

activity

(a)

(b)

strontium
II = 6h

strontium
II = ≈6h

activity

3.5 h

3.5 h

minutes

Figure 11

activity

the 55-day strontium

Figure 12

~54 days

~54 days

~57 days

days →

E

Figure 13

Figure 14

Figure 14a

F

Figure 15
Analysis of the complex nature of 55-day Strontium
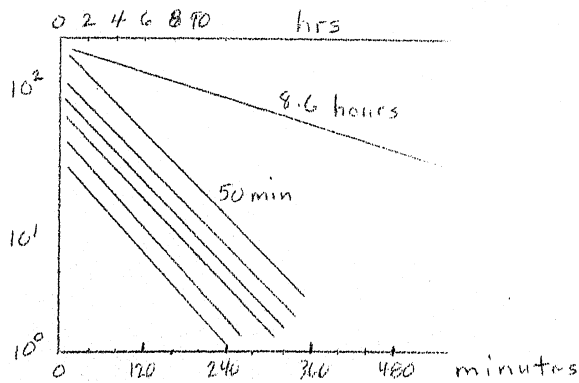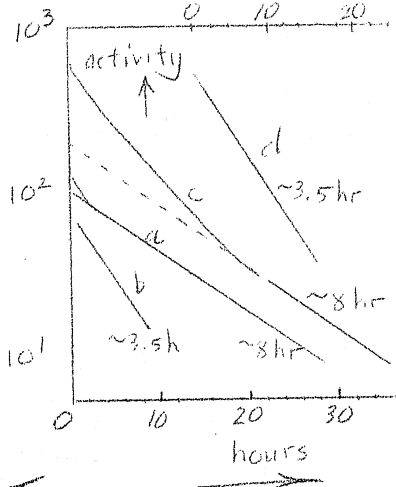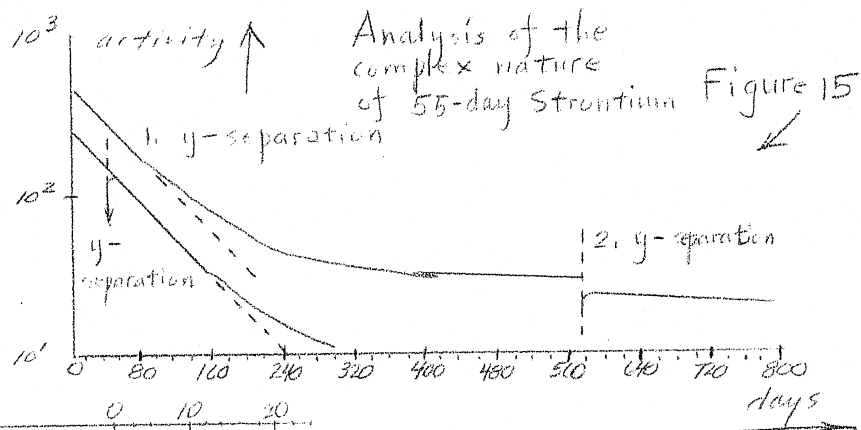

Figure 16
Origin of an 8-hour yttrium from a 7-minute strontium

Figure 17
Analysis of the complex nature of active molybdenum

Suppression of rays
of the 2.3-day substance
in a mixture with the
23-minute uranium by
addition of aluminum foil

Absorption measurements
of element 93

500   1000 μ Al

A

5μ Al

B

5μ Al

100 μ Al

100 μ Al

C

100μ Al

0   40   80   120   160   200

minutes
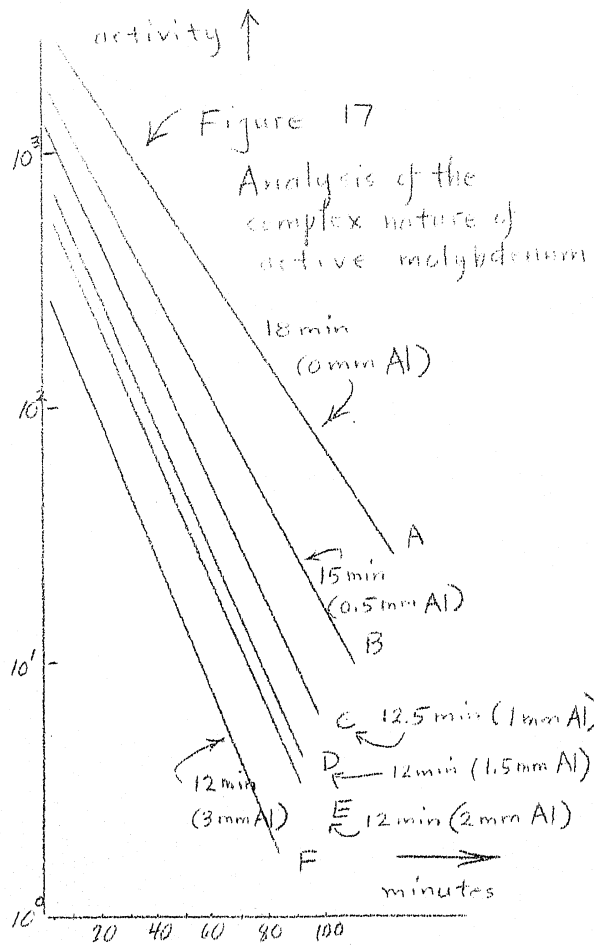
Figure 18

10²

10¹

5   10   15   20
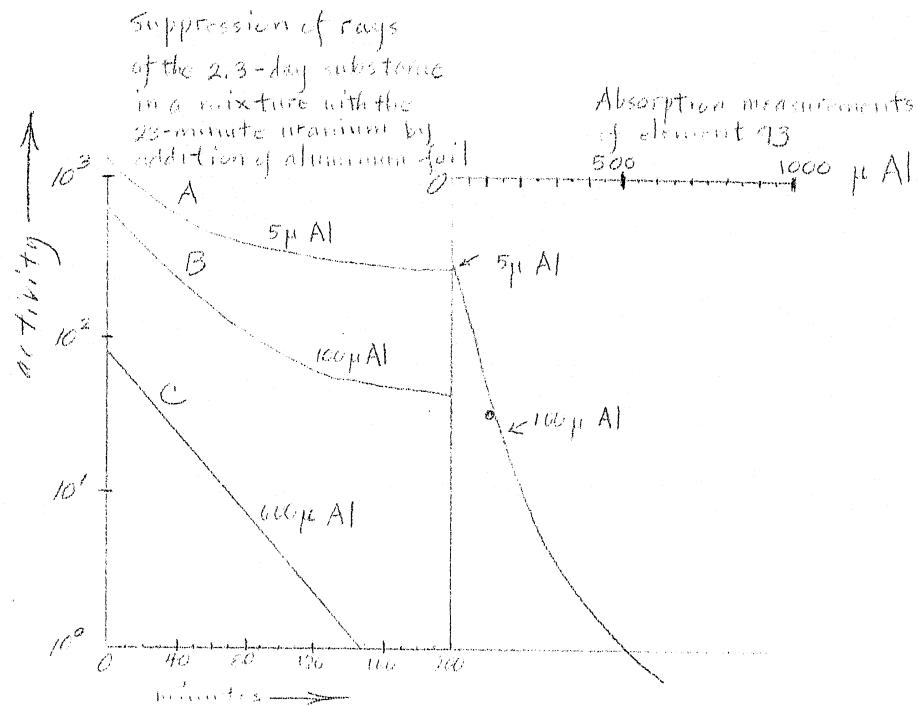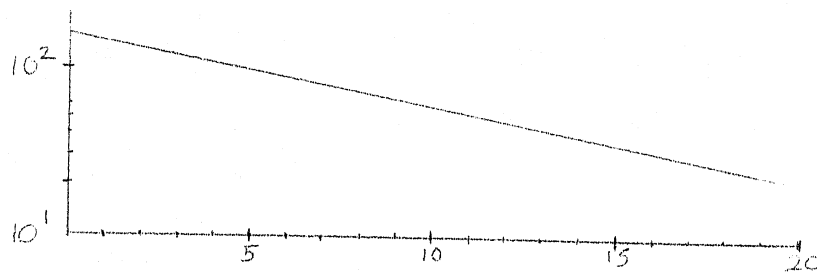
Figure 19   The uranium isotope of 7 day half-life.

H
(END)